

METHOD AND DEVICE FOR THE CATALYTIC REMOVAL OF A POLLUTANT  
CONTAINED IN AN EXHAUST GAS OF A COMBUSTION SYSTEM

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Cross-Reference to Related Application:

This is a continuation of copending international application PCT/DE99/00746, filed March 17, 1999, which designated the United States.

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Background of the Invention:

Field of the Invention:

The invention relates to a method for the catalytic removal of a pollutant contained in an exhaust gas of a combustion system. A predetermined amount of a reagent is thereby introduced into the exhaust gas as a function of the concentration of the pollutant per unit time and is reacted with the pollutant at a catalytic converter. The invention also relates to a device for carrying out the method.

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The combustion of a fossil fuel or of refuse in a combustion system leads to the formation of not inconsiderable quantities of pollutants, such as nitrogen oxides, hydrocarbons, carbon monoxide, sulfur oxide and, in particular, dioxins and furans. 25 These may pass into the environment via the exhaust gas from the combustion system. Examples of a pollutant-emitting

combustion system of this nature include a boiler system, a  
coal-fired, oil-fired or gas-fired fossil power plant, a gas  
turbine, or an internal-combustion engine, in particular a  
diesel engine. Refuse incineration plants also emit the above-  
5 mentioned pollutants.

On account of strict statutory regulations which limit the  
amount of the above-mentioned pollutants which may be  
released, all the above-mentioned combustion systems require  
10 additional treatment of the exhaust gases in order to reduce  
the level of pollutants contained therein. To this end, a wide  
variety of catalytic converters which convert the pollutants  
into compounds which are not hazardous have been developed in  
the past.

15 For example, to reduce the level of pollutants in the exhaust  
gas from a spark-ignition engine (Otto cycle), catalytic  
converters which contain precious metals are known, at which  
hydrocarbons and carbon monoxide together with nitrogen oxides  
20 are converted to form carbon dioxide, nitrogen and/or water.  
Catalytic converters for breaking down dioxins and/or furans  
are also known, comprising a catalytically active material  
based on titanium dioxide. At those catalytic converters the  
dioxins and/or furans are oxidized with residual oxygen which  
25 is present in the exhaust gas to form compounds which are not  
harmful.

If the exhaust gas composition is unsuitable, it may be necessary for an additional reagent to be added to the exhaust gas, which reagent reacts with the pollutant to be eliminated  
5 at a suitable catalytic converter, forming harmless compounds.

By way of example, to break down nitrogen oxides in oxygen-containing exhaust gases, a suitable reducing agent has to be added to the exhaust gas as a reagent. The reducing agent reduces the nitrogen oxides which are present in the exhaust  
10 gas to form harmless nitrogen even in the presence of oxygen. This reaction can be catalyzed by a so-called DeNOx catalytic converter based on titanium dioxide with added vanadium pentoxide, molybdenum trioxide, and/or tungsten trioxide, which uses the selective catalytic reduction process to react  
15 nitrogen oxides with a suitable reducing agent, such as ammonia, to form nitrogen and water.

To fully break down the pollutant in the exhaust gas, the reagent which is added separately must be added in a  
20 stoichiometric amount with respect to the concentration of the pollutant. However, since in combustion systems the concentration of the pollutant in the exhaust gas often fluctuates over time and since, in addition, other factors of the exhaust gas, such as temperature or pressure, influence  
25 the reaction of the pollutant with the reagent, excessive quantities of the reagent are metered in relatively

frequently. This leads to the reagent being emitted into the environment together with the exhaust gas. This emission of reagent, which is also known as reagent slippage or simply slippage, has to be avoided if the reagent itself is harmful.

- 5 In addition, excessive metering of the reagent also always means that operation is uneconomic, since expensive reagent remains unused in the exhaust gas.

Therefore, considerable technical outlay is devoted to

50 attempting to meter the reagent in stoichiometric proportions which are as exact as possible and adapted to the current

state of the exhaust gas. In this context, to determine the amount of reagent which is to be introduced into the exhaust gas per unit time, it is not only necessary for the

15 concentration of the pollutant in the exhaust gas to be either directly measured or calculated on the basis of characteristic

curves derived from relevant operating parameters of the

combustion system, but rather, in addition, it is also

necessary to take into account relevant operating parameters

20 of the catalytic converter, e.g. catalytic converter

temperature, catalytic activity or storage capacity, and

parameters of the exhaust gas, e.g. pressure, gas composition

or temperature, using suitable analysis methods. A technically

complex method of this nature for determining the amount of a

25 reagent metered into an exhaust gas containing nitrogen oxides

is known, for example, from U.S. Patent No. 5,628,186 (see European Patent EP 0 697 062 B1).

However, a complex method of this nature with an expensive  
5 measurement analysis system is inappropriate for retrofitting  
in relatively old combustion systems in particular for cost  
reasons. Also, in particular in the exhaust system of  
internal-combustion engines used to drive vehicles, there is  
no further additional space for sensor elements which are  
10 required to record parameters that are specific to the exhaust  
gas or catalytic converter. In addition, the required  
introduction device has to be of highly dynamic design in  
order to achieve exact metering, so that it can respond  
quickly and accurately under every operating state of the  
15 combustion system with corresponding exhaust gas parameters.  
This also entails excessive costs.

Summary of the Invention:

It is accordingly an object of the invention to provide a  
20 method and device, which overcomes the above-mentioned  
disadvantages of the heretofore-known devices and methods of  
this general type and which provides for a particularly  
inexpensive and simple method for the catalytic removal of a  
pollutant contained in an exhaust gas of a combustion system  
25 by reacting a reagent, without allowing, in the process, any  
significant slippage or excessive consumption of reagent to

occur. It is a further object to also provide a device for carrying out the method which is particularly simple, takes up little space and, in addition, is inexpensive.

5 With the foregoing and other objects in view there is provided, in accordance with the invention, a method of catalytically removing a pollutant contained in an exhaust gas of a combustion system, which comprises:

determining a time average for a concentration of a pollutant

10 in the exhaust gas;

providing a catalytic converter designed for substantially complete conversion of the pollutant given stoichiometric metering of a reagent with respect to the pollutant;

introducing a substoichiometric amount of reagent, in

15 dependence on the time average for the concentration of the pollutant, into the exhaust gas; and

conducting the exhaust gas with the reagent to the catalytic converter and reacting the reagent with the pollutant at the catalytic converter.

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In other words, the time average, or temporal mean, is formed for the concentration of the pollutant, and, in the case of a catalytic converter which, given stoichiometric metering of

the reagent, is designed for substantially complete conversion of the pollutant, the reagent is introduced into the exhaust gas in substoichiometric proportions with respect to the average.

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In a first step, the invention is based on the fact that it is possible to avoid reagent slippage if the pollutant is not converted completely, but rather only to a significant extent.

In fact, in such a case, the reagent can be metered in

10 substoichiometric proportions according to the desired conversion level throughout the entire operating period of the combustion system. This offers sufficient security in the event of minor fluctuations in the concentration of the pollutant in the event of unpredictable operating states of

15 the combustion system. However, with a procedure of this nature the same problem as with stoichiometric metering in any operating state remains. Even in the case of substoichiometric metering, it is necessary to ensure for every operating state of the combustion system that metering is actually taking

20 place in substoichiometric proportions according to the current concentration of the pollutant in the exhaust gas.

Therefore, analytical measurement instruments are once again required in order to prevent slippage of reagent.

25 In a second step, the invention is based on the fact that this outlay can be avoided if it is not each operating state of the

combustion system on its own, but rather the operating period of the combustion system as a temporal medium which is taken into consideration. If a temporal mean for the concentration of the pollutant in the exhaust gas is determined and the  
5 reagent is introduced into the exhaust gas in stoichiometric proportions with respect to the mean, fluctuations in the concentration of the pollutant are absorbed without any additional emission of reagent. If the current concentration of the pollutant in the exhaust gas is  
10 lower than the mean of the concentration which has been determined over a certain period or over a plurality of operating states, the additional amount of reagent is simply reacted with the additional pollutant which is present at the catalytic converter and is consumed in full. Naturally, this  
15 only applies if the catalytic converter is designed in such a manner that substantially complete conversion of the pollutant is achieved if the reagent is metered in stoichiometric proportions with respect to the pollutant. If the current concentration of the pollutant exceeds the mean, the  
20 additional quantity of pollutant remains in the exhaust gas as an unconverted fraction, but over the operating period of the combustion system, this does not diminish the overall conversion rate, which although not complete is still considerable.

The method according to the invention offers the advantage that there is no need for any complex analytical measurement appliances and sensors for monitoring the current exhaust gas composition, for monitoring the current exhaust gas  
5 temperature or for monitoring the current operating state of the catalytic converter, yet reagent slippage is nevertheless reliably avoided. There is also no need to impose any high demands on the introduction device for introducing the reagent into the exhaust gas with regard to the metering accuracy or  
10 the dynamics, since fluctuations in the amount of reagent introduced are absorbed by the fact that the metering is in any case substoichiometric. All this means that the method is predestined for use for exhaust-gas cleaning on old systems or on combustion systems in which, to comply with statutory  
15 exhaust gas regulations, it is not absolutely necessary for all the pollutant in the exhaust gas to be removed. This applies in particular to internal-combustion engines which are used to drive vehicles, in particular to diesel engines, for which the restrictions on pollutant emissions which are  
20 allowable are only being reduced gradually.

In particular, it has been found that slippage of reagent or secondary products which form therefrom can be reliably avoided if the reagent is introduced into the exhaust gas from  
25 the combustion system in substoichiometric proportions with respect to the mean, in such a manner that a proportion of

between 55 and 95% of the pollutant is actually converted. In this case, fluctuations in the emission of pollutant are reliably compensated for without reagent being emitted.

The reagent is advantageously introduced in substoichiometric

5 proportions with respect to the mean, in such a manner that the proportion of the pollutant converted is 75%. In this case, the pollutant is broken down to a considerable extent and, at the same time, there is a sufficient safety margin to compensate for fluctuations in the amount of reagent and/or

10 the concentration of pollutant, without there being any additional emission of reagent downstream of the catalytic converter.

The reagent can be metered in a simple way in that, during an

15 operating state of the combustion system in which the pollutant emission is approximately constant over time, the reagent is also introduced in constant substoichiometric proportions. By way of example, if a mean conversion of the

pollutant of 80% over the operating period of the combustion

20 system is desired, during an operating state of this nature an amount of reagent which is simply reduced by the appropriate factor with respect to the stoichiometric amount is introduced into the exhaust gas according to the concentration of pollutant which is present in the exhaust gas.

In accordance with an advantageous feature of the invention, the concentration of the pollutant may be determined directly by a suitable sensor in the exhaust gas. It is then possible to form a temporal mean from the measured values recorded,

- 5 according to which mean metering is carried out in stoichiometric proportions. The period over which the mean is determined depends on the combustion system. Even in a combustion system with frequent load changes, there is no need to impose high demands on a sensor of this type in terms of  
10 its response speed.

Advantageously, the value for the concentration of the pollutant which is determined by a relatively slow sensor is directly taken into account as a mean for the correspondingly  
15 stoichiometric addition of the reagent.

Alternatively, the concentration of the pollutant in the exhaust gas may be determined on the basis of relevant operating parameters of the combustion system. For this purpose, a corresponding characteristic diagram is stored in a suitable control unit, this diagram representing a functional relationship between an operating state of the combustion system which is defined by the relevant operating parameters and the concentration of the pollutant in the exhaust gas.

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25 Relevant operating parameters may, for example, be the combustion temperature, the level of fuel conversion, the

oxygen consumption or the load. In an internal-combustion engine, the accelerator pedal position, the rotational speed or the torque are also available as relevant operating parameters of this nature.

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The method described is suitable in particular for removing nitrogen oxide using the SCR process. In this case, a suitable reagent is advantageously ammonia or a substance which releases ammonia, in particular urea.

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With the above and other objects in view there is also provided, in accordance with the invention, a device for catalytically removing a pollutant contained in an exhaust gas of a combustion system, comprising:

15 an exhaust pipe conducting an exhaust gas;

an introduction device communicating with the exhaust pipe for introducing a reagent into the exhaust gas;

a catalytic converter configured to conduct a flow of the exhaust gas and for reacting the reagent with a pollutant

20 contained in the exhaust gas, the catalytic converter being  
configured to substantially completely convert the pollutant  
if the reagent is metered into the exhaust gas in  
stoichiometric proportions with respect to a concentration of  
the pollutant; and

a control unit connected to the introduction device for controlling a reagent throughput in the introduction device in dependence on the concentration of the pollutant in the exhaust gas, and for determining a time average of the 5 concentration of the pollutant in the exhaust gas and for metering in the reagent substoichiometrically according to the time average.

In other words, to carry out the method, the invention

10 provides a device for the catalytic removal of a pollutant contained in an exhaust gas of a combustion system, having an exhaust pipe, having an introduction device for introducing the reagent into the exhaust gas, having a catalytic converter through which the exhaust gas/reagent mixture can flow, for 15 reacting the reagent with the pollutant, and having a control unit for controlling the reagent throughput in the introduction device as a function of the concentration of the pollutant in the exhaust gas, in which device, according to the invention, the catalytic converter is designed in such a 20 manner that, if the reagent is metered in stoichiometric proportions with respect to the concentration of the pollutant, the pollutant is substantially completely converted, and in which device the control unit is designed to determine a temporal mean of the concentration of the 25 pollutant in the exhaust gas and for substoichiometric metering according to the mean. Advantageously, the control

unit is designed for substoichiometric metering, in such a manner that the proportion of the pollutant which is converted at the catalytic converter is between 55 and 95%, preferably 75%. For this purpose, the control unit records the current 5 value of the concentration of the pollutant in the exhaust gas and, from this value, determines a mean of the concentration which is determined over a certain period. Then, a correspondingly substoichiometric amount of reagent is introduced into the exhaust gas via the introduction device 10 using a predetermined reduction factor with respect to the stoichiometric amount.

In a combustion system which is operated with frequent load changes, it is for this purpose advantageously possible for 15 the control unit to recognize operating states in which the emission of pollutant is virtually constant and to meter in substoichiometric proportions of reagent during this period in a suitable way. All that is required is for the desired mean conversion of the pollutant to be between 55 and 95%, 20 preferably 75%.

The introduction device itself may in a known way be in the form of an injection valve, an injection nozzle or, in the case of large systems, in the form of a spray grid distributed 25 over the cross section of the exhaust pipe. The metering may be effected either by direct control of the injection nozzle

or the injection valve or via a controllable valve provided in the feed line for the reagent.

To record the concentration of the pollutant in the exhaust gas, a suitable sensor is advantageously arranged in the exhaust line between the combustion system and the catalytic converter. If a long response time means that the sensor takes a mean over various operating states of the combustion system, there is no need for the control unit itself to recognize

operating states in which the emission of pollutant is approximately constant. It is merely necessary for the value of the concentration of the pollutant determined by means of the sensor to be taken into account for stoichiometric metering.

In accordance with another advantageous configuration of the invention, the concentration of the exhaust gas is determined on the basis of relevant operating parameters of the combustion system. For this purpose, as described above, a

corresponding characteristic diagram or characteristic curve is stored in the control unit. To determine the relevant operating parameters, the control unit is assigned suitable sensor elements. This procedure is recommended in particular in an internal-combustion engine which has an electronic engine management system and therefore a suitable electronic interface. Information concerning the operating state of the

combustion system, such as for example combustion temperature, fuel consumption, torque, rotational speed, as well as injection time and the like, can be extracted easily via an interface of this type. The sensors provided for this purpose 5 form part of the prior art.

Particularly for the removal of nitrogen oxides using the SCR process, it is advantageous if the catalytic converter is designed as a DeNOx catalytic converter in honeycomb form and 10 comprises the materials titanium dioxide in a proportion of 70 to 95% by weight, tungsten trioxide and/or molybdenum trioxide in a proportion of 5 to 20% by weight, and vanadium pentoxide in a proportion of less than 5% by weight. For this purpose, the reagent used may be ammonia or a substance which releases 15 ammonia, such as urea.

Other features which are considered as characteristic for the invention are set forth in the appended claims.

20 Although the invention is illustrated and described herein as embodied in a method and device for the catalytic removal of a pollutant contained in an exhaust gas of a combustion system, it is nevertheless not intended to be limited to the details shown, since various modifications and structural changes may 25 be made therein without departing from the spirit of the

invention and within the scope and range of equivalents of the claims.

The construction and method of operation of the invention,  
5 however, together with additional objects and advantages thereof will be best understood from the following description of specific embodiments when read in connection with the accompanying drawing.

10 Brief Description of the Drawing:

The figure of the drawing is a schematic diagram of a diesel engine with a connected device for the catalytic removal of the nitrogen oxides from the exhaust gas.

15 Description of the Preferred Embodiments:

Referring now to the sole figure of the drawing in detail, the exemplary combustion system shown therein is a diesel engine 1 which is fitted with an exhaust-gas cleaning device for eliminating nitrogen oxides. The diesel engine 1 has an interface 3 from which the current values of relevant operating parameters can be derived by electronic means. A fuel/air mixture for combustion is supplied to the diesel engine via a fuel feed 4 and an air intake 5. The exhaust gas 6 from the diesel engine is passed via a manifold into an exhaust pipe 7 and is discharged to the outside via a catalytic converter 8. In this case, the catalytic converter 8

is a so-called DeNOx catalytic converter which uses the known SCR (selective catalytic reduction) process to break down nitrogen oxides with the aid of the reagent ammonia to form molecular nitrogen and water. The amount of ammonia required 5 is obtained by hydrolysis from urea which is metered in.

To meter the urea, an introduction device 9 is provided, which comprises a tank 11 for the reagent 10 (= urea), a feed line 12, a metering valve 13 and an injection nozzle 14.

10 The catalytic converter 8 for breaking down the nitrogen oxides is designed as an unsupported extrudate in honeycomb form made from a ceramic mass comprising 90% by weight titanium dioxide, 8% by weight tungsten trioxide and less than 15 2% by weight vanadium pentoxide. The catalytic converter 8 is designed in such a manner that even when the maximum level of nitrogen oxides is being emitted, the nitrogen oxides are substantially completely broken down if the reagent 10 is metered in in stoichiometric proportions.

20 In addition, a commercially available pollutant sensor 15 for recording the nitrogen oxide concentration as a result of a change in conductivity is arranged in the exhaust pipe 7 between the combustion system 1 and the catalytic converter 8.

To control the introduction device, a control unit 18 is provided. The current values for fuel consumption, speed and torque are available to the control unit via the outputs 19, 20 and 21 of the interface 3 of the diesel engine.

- 5 Furthermore, it is possible to access the measured nitrogen oxide emissions from the diesel engine via the output 22 of the pollutant sensor 15.

It is optionally possible for the nitrogen oxide concentration, which is determined via the pollutant sensor 15 or is calculated using a characteristic curve which is implemented in the control unit 18 on the basis of the relevant operating parameters fuel consumption, speed and torque, to be used directly in order to determine the amount 15 of reagent 10 which is to be introduced per unit time.

In operating states of the combustion system in which the concentration of the nitrogen oxides in the exhaust gas is approximately constant over time, the control unit 18 controls 20 the metering valve 13 of the introduction device 9 in such a manner that an amount of reagent 10 which is reduced correspondingly according to a predetermined reduction factor with respect to the stoichiometric amount of reagent 10 is added to the exhaust gas 6 per unit time. The urea 25 introduced breaks down in the exhaust gas 6 as a result of hydrolysis, forming ammonia and residues. The ammonia reacts

as a reagent with the nitrogen oxide at the catalytic converter 8. For the control unit, it is possible to input a conversion level for the pollutant of between 55 and 95%.

- 5 In the case where the concentration of the pollutant is calculated from relevant operating parameters, the control unit 18 records an average over a period of minutes. In the case where the concentration is determined by means of the pollutant sensor 15, the directly measured value is used as a
- 10 mean.

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